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Atomic Resolution Imaging of $YAlO_3$:Ce in the Chromatic and Spherical Aberration Corrected PICO Transmission Electron Microscope

Lei Jin[1], Juri Barthel[1,2], Chun Lin Jia[1,3] and Knut W. Urban[1,3]

[1.] Ernst Ruska-Center for Microscopy and Spectroscopy with Electrons, Jülich-Aachen Research Alliance (JARA), Forschungszentrum Jülich GmbH, 52425 Jülich, Germany.
[2.] Central Facility for Electron Microscopy, RWTH Aachen University, 52074 Aachen, Germany.
[3.] School of Electronic and Information Engineering and State Key Laboratory for Mechanical Behavior of Materials, Xi'an Jiaotong University, Xi'an 710049, China.

What is the real resolution that can be realized in practical electron microscope operation? We have investigated this problem for TEM imaging in a systematic study on Ce-doped yttrium orthoaluminate, $YAlO_3$. Employing a FEI Titan G3 50-300 (PICO) electron microscope equipped with the new CEOS C-COR+ corrector for spherical (C_S) and chromatic aberration (C_C) we studied the conditions to resolve the 57 pm Y-Y-atom pair distance in projection along [010]. Our successful imaging of this pair proves a record resolution for direct 200 kV TEM imaging [1].

In a C_S and C_C corrected electron microscope the two major resolution limitations of previous instrument generations due to partial spatial and partial temporal coherence are significantly reduced. The spatial resolution of such an instrument is limited by an incoherent image spread [2]. This dampens linear and non-linear contributions at the same image frequency by an equal amount. This is an essential qualitative difference compared to the previously dominating partial temporal coherence effects, which in general cause a different dampening of linear and non-linear contributions. Besides the typical sources of image spread, such as mechanical vibrations of the specimen holder and electrical instabilities of beam deflectors, thermal magnetic field-noise, also known as Johnson noise, is currently considered as the major contribution to the image spread in the C-COR setup [3]. The C-COR allows to effectively reduce unwanted coherent axial aberrations of the imaging system up to the fourth order (C_4), and to obtain optimum phase-contrast transfer up to the information limit by adjusting also the fifth-order spherical aberration (C_5). The PICO microscope is equipped with an advanced version, C-COR+, of the image corrector. It is optimized for an as low as possible level of Johnson noise thus providing an improved resolution compared to that of C-COR [3] at 200 kV.

In order to allow a comparison of the performance of different instruments resolution has inevitably to be defined universally and independent of sample parameters. In reality, however, these parameters may be decisive for the *direct* perception of resolution in practical work. On the other hand, today, where information limits in the range of 50 pm are characterizing electron microscopy, the dilemma of an insufficient image-signal separation due to sample parameters applies quite frequently and contrast simulations have become compulsory for image interpretation. These calculations are providing also an indirect access to the value of a microscope's spatial resolution. However, a necessary condition in order to be able to extract the instrument resolution based on image simulations is a quantitative comparison of experimental intensities with calculated values *on the same absolute scale*. This has just recently been achieved [4]. Within this numerical forward modeling of the imaging process in TEM the *real* atomic separation in the sample can be determined irrespective of whether this is directly visible in the images.

Optimum contrast transfer for NCSI conditions considering third-order (C_3) spherical aberration and C_5

was treated in [5]. Negative phase contrast, providing bright image contrast at atomic positions against a dark background, is obtained over a certain range of sample thicknesses for a negative value of C_3 combined with a positive value of C_5 and a positive defocus value C_1 (overfocus). Our NCSI work was accompanied by detailed state-of-the art image simulations. These confirmed an experimental resolution of 52 pm. Images were recorded employing 4k × 4k Gatan UltraScan 4000 UHS camera at a sampling rate of 4.3 pm/pixel.

Figure 1 displays an experimental image. The schematic (upper left corner) outlines an area of 2 × 2 unit cells. The Y-Y-atom pairs (two orientations) and the oxygen atoms are resolved with high contrast. Frequently one of the two Y-atom columns forming a pair shows stronger contrast compared to the other. As confirmed by contrast simulations this indicates a difference of ± 1 atom in length of the two columns. Figure 2 shows that the atom-pair distances vary between 55 pm and 80 pm (mean value: 68.9 pm; standard deviation: 7.8 pm). This is attributed to lattice relaxation in the thin sample (1.7 nm) used. We note that the 57 pm atom-pair separation is clearly resolved for both orientations. To the best of our knowledge this demonstrates the highest direct resolution in coherent TEM atomic imaging in materials at 200 kV to date. Finally we would like to acknowledge an earlier brief report on $YAlO_3$:Ce imaging by HAADF STEM [6]. Coherent TEM and incoherent STEM with 200 keV electrons show here comparable resolving power, as the close Y-Y separation can be directly observed in images obtained by either technique. However, in contrast to STEM, imaging by NCSI TEM shows also the oxygen atoms.

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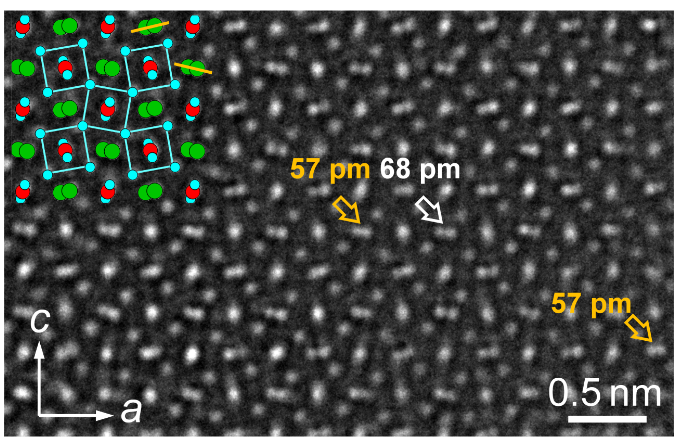

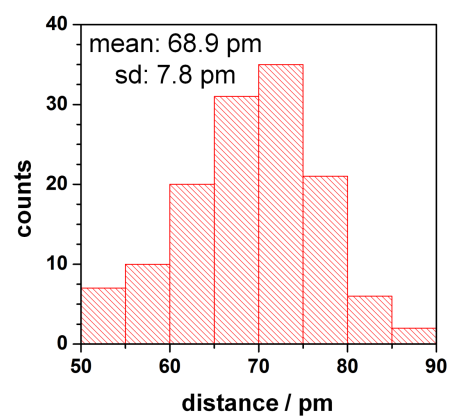

Figure 1 (left). Image of $YAlO_3$:Ce in [010] projection. Insert: Outline of 2 × 2 unit cells. Corners of squares give position of O atoms (blue); centers denote positions of Al atoms (red) overlapping with O atoms; Y atoms green. Arrows denote examples of Y-Y- atom pairs with pair separations given.

Figure 2 (right). Distribution of atom-pair separations in the image determined from Gaussian fits; “sd” denotes the standard deviation of the data.

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